

Program LMTART for Electronic Structure Calculations

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A computer program LMTART for electronic structure calculations using full potential linear muffin-tin orbital method is described.

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I. DESCRIPTION

Full-potential linear-muffin-tin-orbital (Ref.¹) (FP-LMTO) program LMTART is designed to perform band structure, total energy and force calculations of solids using methods of density functional theory (DFT) (Ref.²). The development of LMTART has been initiated in 1986 in P.N. Lebedev Physical Institute. It was further extensively developed in Max-Planck Institut für Festkörperforschung, and its most recent contributions have been done in Departments of Physics of Rutgers University and of New Jersey Institute of Technology.

LMTART performs electronic structure calculations using popular local density approximation (LDA) and generalized gradient approximation (GGA) of density functional theory². To deal with strongly correlated systems it also implements LDA+U method³ and will include the capability to study materials using spectral density functional approach and dynamical mean field theory (DMFT)⁴ in the nearest future. For determining equilibrium crystal structure and phonon spectra LMTART computes total energies, forces⁵ as well as full wavevector dependent lattice dynamics⁶ using linear response theory. For studying optical spectral functions LMTART evaluates dipole matrix elements between various electronic states and can determine optical conductivity of materials.

The code implements atom centered local muffin-tin orbitals as a basis for representing one-electron wave functions in both bare and screened (tight-binding) representations with general choice of tail energies κ^2 . This allows to increase accuracy in the calculations by using multiple- κ LMTO expansions. Low lying core electronic states can be resolved as bands in separate energy panels. For heavy elements, spin-orbit coupling matrix elements can be taken into account using a variational procedure proposed by Andersen¹. Full three dimensional treatment of magnetization in relativistic calculations including LDA+U is implemented to study problems related to magnetic anisotropy and non-collinear magnetism. Effect of finite temperature can be modelled by introducing Fermi statistics for the one-electron states. Additionally, LMTART can determine tight-binding fits to the energy bands by extracting hopping integrals between various orbitals using tight-binding LMTO representation.

LMTART is written on FORTRAN 90 and uses dynamical memory scheme. No additional recompilation of the code is required when changing numbers of atoms,

spins, plane waves, etc. There are two basic regimes for working with this program: the self-consistent charge density calculation and calculation of physical properties such as electronic structure, optical properties, etc.. The simplest input to LMTART involves only atomic charges of the atoms as well as crystal structure.

LMTART works with two different approximations related to a shape of the potential: (i) atomic sphere approximation (ASA) and plane wave expansion (PLW). ASA uses overlapping atomic spheres, where the potential is expanded in spherical harmonics inside the spheres, but any contribution from the interstitial region is neglected. Such method is fast and provides reasonably good energy bands. However it is not sufficiently accurate to deal with distortions and phonons. PLW is a full potential approximation which uses non-overlapping muffin-tin spheres, where the potential is represented via spherical harmonics expansions, and the interstitial region where the potential is expanded in plane waves. The full potential regime provides the best accuracy at the price of increasing computational time. Finally, LMTART can be run in a special tight-binding regime by supplying hopping integrals between various orbitals.

Two extensions of LMTART are available as separate programs. One is a full-potential linear-response linear-muffin-tin-orbital package LMTO PHONONS which is designed to perform linear-response calculations of the phonon spectra for arbitrary wave vectors \mathbf{q} . Main features of this code include: (i) Computations of first-order changes in the charge density and the potential due to displacements of nuclei for arbitrary wave vectors \mathbf{q} . (ii) Calculations of the dynamical matrix and the phonon spectra. (iii) Calculations of electron-phonon interactions, Eliashberg spectral functions $\alpha^2F(\omega)$ and transport properties such as electrical and thermal resistivities. The description of the method and complete set of references can be found in Ref.⁶. Numerous applications are given in Ref.⁷.

Also available is an extension of LMTART to compute dynamical susceptibilities and magnon spectra. This is a full-potential linear-response linear-muffin-tin-orbital package of programs LMTO MAGNONS designed for these purposes. As a by-product, spin wave spectra and their lifetimes are accessible as peaks in calculated imaginary spin susceptibility. A short description of the method can be found in Ref.⁸.

Software MINDLab which runs under Microsoft Windows operating systems can be used to set up input files

and analyze output files of LMTART. It can also be used to run the code without learning its extensive input options. This software is necessary to visualize all data withdrawn from LMTART. Currently, MINDLab can perform the following tasks (i) Crystal group calculations. By setting atomic positions MINDLab shows crystal group operations found for given atomic configuration; (ii) Fat bands calculations. Fully colored visualization of separated orbital characters on top of the band structure can be performed by mouse click operations. (iii) Density of states calculations. MINDLab computes and visualizes densities of states, total and orbital resolved. (iv) Extractions of hopping integrals for tight-binding fits can be performed quickly with MINDLab. (v) Optical properties calculations. Dielectric functions $\epsilon_1(\omega)$, $\epsilon_2(\omega)$, and electron energy loss spectra can be computed and visualized by MINDLab. (vi) Visualizations of charge densities, full potentials, and Fermi surfaces in both two and three dimensions using OpenGL graphical library implemented within MINDLab (viii) Three dimensional visualization of crystal structures and their distortions. (ix) Full three dimensional visualization of vector fields including velocity fields on top of the Fermi surfaces and magnetizations. (x) Simplified input for studying correlated electronic systems which currently implements LDA+U method and will include LDA+DMFT technique in the nearest future.

MINDLab works with LMTART as with an executable file which reads input data, performs band structure calculation, and stores output files. MINDLab controls this process, it prepares input for LMTART using dialog windows, and executes LMTART program as a separate thread. When the calculation is finished, LMTART notifies the MINDLab, and the output files can be visualized by simple mouse click operations. While running LMTART for complicated compounds can be rather slow (it depends how many atoms per unit cell is selected), MINDLab can be used to prepare the input, after which the files can be copied to a more powerful computer where the LMTART can be executed much faster. After run is performed, the output can be copied back to the PC and analyzed quickly by MINDLab. All input/output files of LMTART are formatted ASCII files, and therefore system independent.

II. APPLICATIONS

Several most recent applications of LMTART can be found in Refs.^{4,7,8,9,10}. They include calculations of phonons and electron-phonon interactions in novel superconductors, such as MgB_2 , LiBC , MgCNi_3 , studies of phonons in strongly correlated systems, e.g. NiO , Pu , computations of magnetic anisotropy energies of ferromagnets Fe , Co , Ni , CrO_2 evaluations of dynamical spin susceptibilities and spin wave spectra, and so on.

Here we illustrate one of the most recent calculation⁹ of phonon spectrum in MgCNi_3 using a linear response

regime of LMTART. The calculated phonon dispersions along major high symmetry lines of the cubic Brillouin zone are given on Fig. 1. The frequencies are seen to be span up to 1000 K, with some of the modes showing significant dispersion. In general, we distinguished three panels where the top three branches around 900 K are carbon based, the middle three branches around 600 K are Mg based and 9 lower branches are all Ni based.

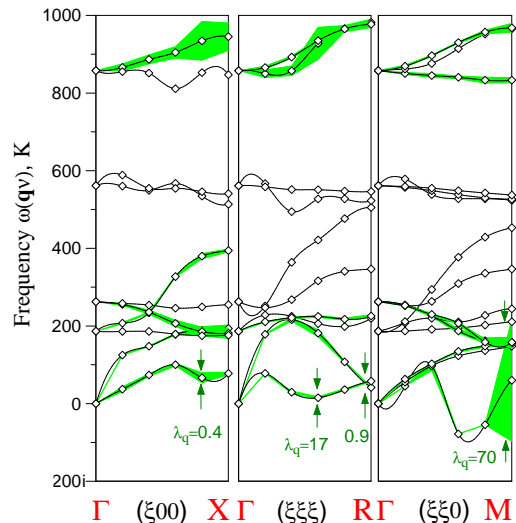


FIG. 1: Calculated phonon spectrum of MgCNi_3 along major symmetry directions of the Brillouin zone of the cubic lattice using density functional linear response method. Some curves are widened proportionally to the phonon linewidths.

We have discovered a striking feature of this phonon spectrum connected to the presence of a low-frequency acoustic mode which is very soft and is even seen to be unstable along some points in $(\xi\xi0)$ direction in the Brillouin zone. This mode is essentially Ni based and corresponds to perpendicular movements of two Ni atoms towards octahedral interstitials of the perovskite structure. The latter is made of each of the four Ni atoms and two Mg atoms.

The persistence of the instability which does not occur for any of the high-symmetry point needs a non-trivial frozen-phonon analysis. As our polarization vectors prompt that the largest anharmonicity is expected for the ΓXM plane ($q_z = 0$) of the Brillouin zone of the cubic lattice, we have performed three such calculations for the points $a = (\frac{1}{4}\frac{1}{4}0)\frac{2\pi}{a}$, $b = (\frac{1}{2}\frac{1}{4}0)\frac{2\pi}{a}$, and M . The results of these calculations reveal essentially anharmonic interatomic Ni potentials. A shallow double well with a depth of the order of 20–40 K and the curvature at the equilibrium of the order of 40–50i K exists for the a and b points which becomes vanishing at the M point. We have found such a behavior by both total energy and force calculations for the supercells of 20 atoms (a and b points) and 10 atoms (M point). Clearly, such a small depth on the temperature scale indicates that the distortions are dynamical and zero point motions would prevent the

appearance of the static long-range order. Complete discussion of these calculations as well as the comparisons with existing experiments can be found in Ref.⁹.

Fig.1 also shows the calculated phonon linewidths $\gamma_{\mathbf{q}\nu}$ by widening some representative phonon dispersion curves $\omega_{\mathbf{q}\nu}$ proportionally to $\gamma_{\mathbf{q}\nu}$. In particular, we found that some phonons have rather large linewidths. This, for example holds, for all carbon based higher lying vibrational modes. The strength of the coupling, $\lambda_{\mathbf{q}\nu}$, for each mode can be obtained by dividing $\gamma_{\mathbf{q}\nu}$ by $\pi N(\epsilon_F)\omega_{\mathbf{q}\nu}^2$, where $N(\epsilon_F)$ is the density of states at the Fermi level equal to 5.3 st./[eV*cell] in our calculation. Due to large $\omega_{\mathbf{q}\nu}^2$, this unfortunately results in strongly suppressed coupling for all carbon modes which would favor high critical temperatures. The coupling, however, is relatively strong for the Ni based modes. For example, we can find λ 's of the order of 1–3 for the Ni based optical phonons around 250 K. Our resulting value of electron-phonon coupling constant λ is found 1.51. This assumes that MgCNi₃ is a strongly coupled electron-phonon superconductor.

III. DOWNLOADS

All LMTART based programs described above are available for downloading at the following URL:

<http://www.physics.njit.edu/~mindlab>. Each program consists of the source code, manual, and example files. An extensive database of electronic structures for different materials is also available at this HTTP URL.

IV. ACKNOWLEDGEMENTS

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